LBNL-47531 *59th Ann. Proc. MSA, Long Beach, California (2001) 898-899.*

SUB-ÅNGSTROM TRANSMISSION ELECTRON MICROSCOPY AT 300keV

M.A. O'Keefe*, E.C. Nelson*, J.H. Turner* and A. Thust°

*National Center for Electron Microscopy, LBNL B72, Berkeley, CA 94720, USA
°Institut für Festkörperforschung, Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany

SUB-ÅNGSTROM TRANSMISSION ELECTRON MICROSCOPY AT 300keV

M.A. O'Keefe*, E.C. Nelson*, J.H. Turner* and A. Thust°

*National Center for Electron Microscopy, LBNL B72, Berkeley, CA 94720, USA
°Institut für Festkörperforschung, Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany

Sub-Ångstrom TEM to a resolution of 0.78Å has been demonstrated by the one-Ångstrom microscope (OÅM) project at the National Center for Electron Microscopy. The OÅM combines a modified CM300FEG-UT with computer software[1,2] able to generate sub-Ångstrom images from experimental image series.

Sub-Ångstrom HREM is gaining in importance as researchers design and build artificially-structured nanomaterials such as semiconductor devices, ceramic coatings, and nanomachines. Commonly, such nanostructures include atoms with bond lengths shorter in projection than the point resolution of a mid-voltage HREM[3]. In addition, image simulations have shown that structure determinations of defects such as dislocation cores require sub-Ångstrom resolution[4], as will hold true for grain boundaries and other interfaces.

Sub-Ångstrom microscopy with a transmission electron microscope requires meticulous attention to detail. As resolution is improved, resolution-limiting parameters need to be reduced. In particular, aberrations must be minimized, power supplies must be stabilized, and the microscope environment optimized to reduce acoustic and electromagnetic noise in addition to vibration. Figure 1 shows limits for several important parameters. To reach a direct resolution of d_S the spherical aberration coefficient C_S needs to be below $6d_S^4/\lambda^3$. Thus, to reach 0.8Å at 300keV (fig.1), C_S should be less than 0.03mm (0.02mm would be optimum[5]). Alternatively, to reach an information limit of d_Δ by focal reconstruction requires the standard deviation of focus spread Δ to be less than $2d_\Delta^2/(\pi\lambda)$, or 21Å at 300keV for d_Δ=0.8Å. Two- and three-fold astigmatism, A_1 and A_2 must be kept low. To ensure phase distortions of less than $\pi/4$ at resolutions of d_{A_1} and d_{A_2}, A_1 and A_2 must be below $d_{A_1}^2/(4\lambda)$ and $3d_{A_2}^3/(8\lambda^2)$ respectively, or 8Å and 500Å to reach 0.8Å at 300keV. Even specimen thickness must be reduced[6] to less than $2d_S^2/\lambda$, requiring less than 65Å to reach 0.8Å at 300keV.

Sub-Ångstrom resolution was achieved with the OÅM by placing the TEM in a favorable environment[7], and by reducing its three-fold astigmatism A_2, and information limit d_Δ. Before correction, A_2 was measured as 2.46μm (fig.2a); after correction, as 300Å (fig.2b), corresponding to 0.68Å at a $\pi/4$ phase limit. Measurement of the energy spread (gun spread plus high-voltage ripple) as 0.93eV FWHH indicated a focus spread Δ of 20Å and an information limit of 0.78Å (fig.1). Tests with a diamond specimen showed that A_2 was corrected and the OÅM could successfully resolve the 0.89Å (400) dumbbell spacings in [110] diamond.[3,8,9]

Sub-Ångstrom resolution is improved by lowering the TEM information limit. Measurements showed that the expected limit could be lowered below 0.75Å by reducing the gun extraction voltage (fig.3). As a test, we have imaged the 0.78Å (444) dumbbell spacings in [112] silicon. The 444 reflections have optimum transfer into the image at an alpha-null defocus[10] of -3783Å (fig.4). Operating with reduced illumination at 3kV extraction voltage, we obtained focal series of images that produced diffractogram spots out to 0.78Å (fig.5). In 10Å-step focal series, the 0.78Å (444) spots showed the expected Fourier period[11] of 62Å, reaching maximum intensity every 60 to 70Å as predicted (fig.4), indicating linear transfer from the specimen.

Sub-Ångstrom spacings of 0.78Å appear in our focal series. At the extreme limit of the microscope's performance, images are noisy, but 0.78Å dumbbells appear at both "white-atom" and "black-atom" defocus, indicating linear transfer of the 0.78Å spacing from the specimen. A distinct pair of "white atoms" is clearly seen in the top left of fig.6a. "Black-atom" separation is best revealed when the image is profiled (fig.6b).[12]

1. W.M.J. Coene, A. Thust, M. Op de Beeck and D. Van Dyck, *Ultramicroscopy* **64** (1996) 109-135.
2. A. Thust, W.M.J. Coene, M. Op de Beeck and D. Van Dyck, *Ultramicroscopy* **64** (1996) 211-230.
3. Y.C. Wang et al., *57th Ann. Proc. MSA,* Portland, Oregon (1999) 822-823.
4. C. Kisielowski et al., in *58th Ann. Proc. MSA*, Philadelphia, Pennsylvania (2000) 16-17.
5. Michael A. O'Keefe, in *58th Ann. Proc. MSA*, Philadelphia, Pennsylvania (2000) 1036-1037.
6. Michael A. O'Keefe, in *55th Ann. Proc. MSA,* Cleveland, Ohio (1997) 1165-1166.
7. John H. Turner, Michael A. O'Keefe & Robert Mueller, *55th Ann. Proc. MSA,* Cleveland, Ohio (1997) 1177-1178.
8. Michael A. O'Keefe, in *58th Ann. Proc. MSA*, Philadelphia, Pennsylvania (2000) 1192-1193.
9. Michael A. O'Keefe et al., *Ultramicroscopy* (2001) submitted.
10. Michael A. O'Keefe, these proceedings.
11. Sumio Iijima and Michael A. O'Keefe, J. Micros. **117** (1979) 347-354.
12. Work supported by the Director, Office of Science -- through the Office of Basic Energy Sciences, Material Sciences Division, of the U.S. Department of Energy, under contract No. DE-AC03-76SF00098.

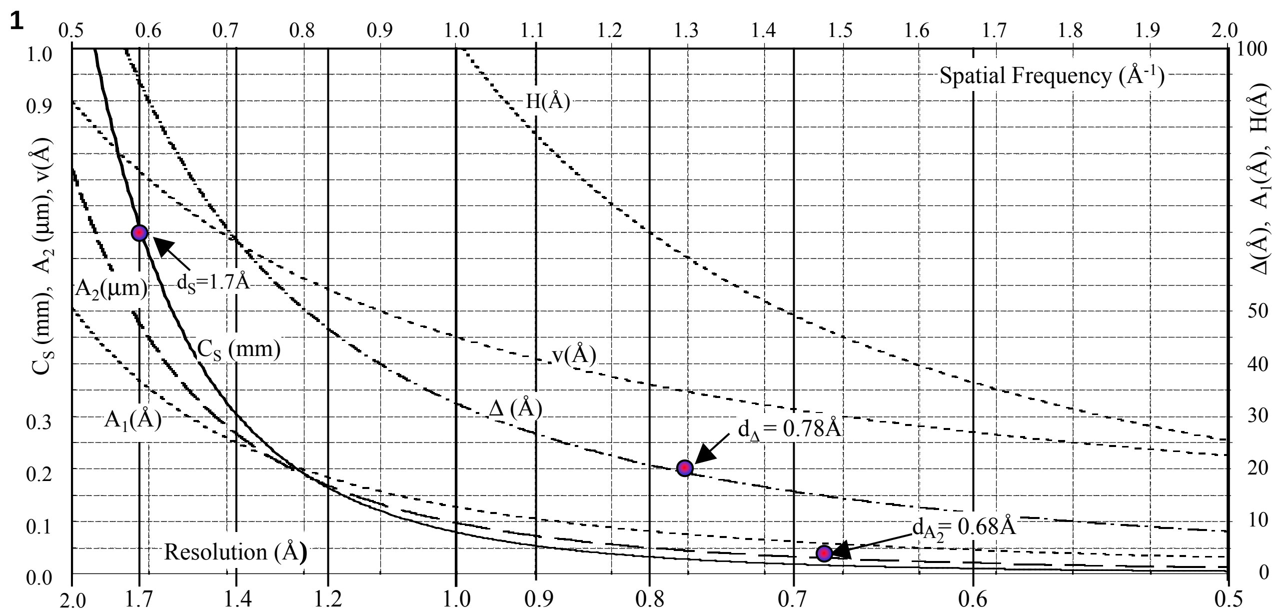


Fig.1 Resolution-limiting parameters plotted for spatial frequency (top) from 0.5 to 2.0Å^{-1} and resolution from 2.0 to 0.5Å (bottom) for 300keV. Spherical aberration C_S(mm), 3-fold astigmatism A_2(μm) and vibration v(Å) are plotted from 0 to 1 (left). Spread of focus Δ, 2-fold astigmatism A_1, and specimen thickness H range from 0 to 100Å (right).

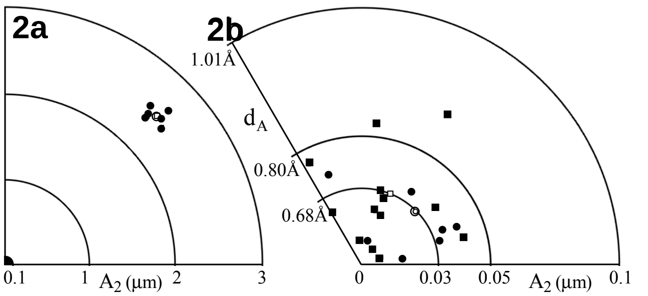


Fig.2. Measured 3-fold astigmatism before (a) and after (b) correction. Mean values are hollow. Mean in (a) is 2.46μm. Means in (b) are 0.03μm, corresponding to π/4 at 0.68Å.

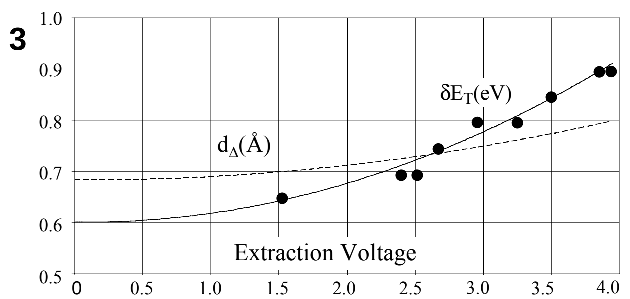


Fig.3. As extraction voltage is reduced from 4.0keV, measured FWHH energy spread δE_T(eV) falls from 0.93eV, and information limit d_Δ(Å) falls from 0.8Å.

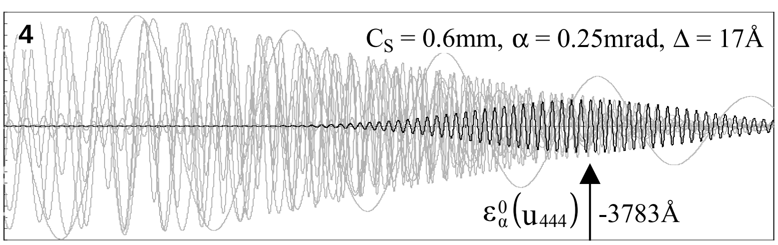
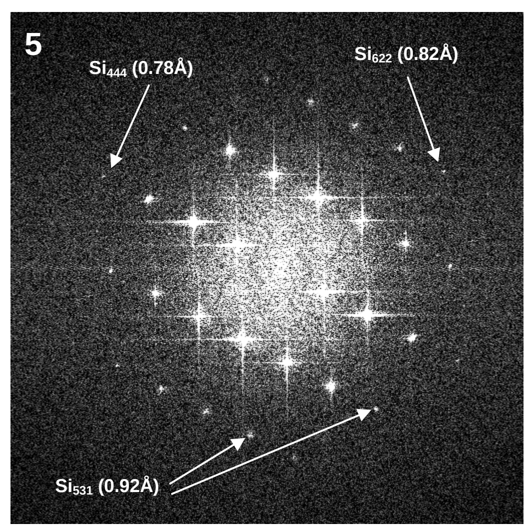


Fig.4. Plot of beam phases shows position of α-null defocus for 0.78Å.



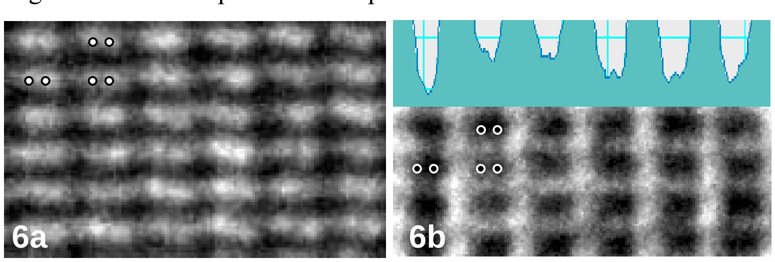


Fig.6. Images of Si[112] show separation of 0.78Å Si-Si dumbbells. (a) “white-atom” image. (b) “black-atom” dumbbell is difficult to see, but profile (averaged vertically over 6 dumbbells) shows separation.

Fig.5 Diffractogram from Si[112] image has intensities that exhibit linear-transfer Fourier-defocus behavior down to 0.78Å.